

AN UNSTEADY TWO-DIMENSIONAL ANALYTICAL SOLUTION FOR MODELING AIR POLLUTION DISPERSION AND TURBULENT DRY DEPOSITION

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***Abstract.** Dry deposition at the surface of air pollution from a ground-level or an elevated source can be considered in different ways. The deposition flux is usually parameterized in terms of deposition velocity, which is either specified empirically or estimated from appropriate theoretical relations. Using the gradient transport, dry deposition is included by specifying the deposition flux as the surface boundary condition. The advection-diffusion equation can be written in finite difference form, thus opening the door to a countless variety of numerical solutions. Analytical solutions of equations are of fundamental importance in understanding and describing physical phenomena. Analytical solutions (as opposed to numerical ones) explicitly take into account all the parameters of a problem, so that their influence can be reliably investigated and sensitivity analysis over model parameters may be easily performed. Moreover, numerical codes based on analytical expressions need less computational resources. In this work we present a general solution (i.e. for any wind and eddy diffusivity vertical profiles) for the unsteady two-dimensional advection-diffusion equation with dry deposition to the ground. The above solution is obtained applying Generalized Integral Laplace Transform Technique (GILTT). Moreover, we will report numerical simulations of the ground-level concentrations compared with an experimental data set.*

Keywords: GILTT, Dry Deposition, Advection-Diffusion Equation, Analytical Solution

1. INTRODUCTION

The advection-diffusion equation can be written in finite difference form, thus paving the way to a countless variety of numerical solutions. Using the gradient transport approach (K-theory), dry deposition is included by specifying the deposition flux as the surface boundary condition. Therefore, numerical solutions to the advection-diffusion equation with variable eddy diffusivities are used to take into account the effects of dry deposition as well as gravitational settling for heavier particles (Arya 1999).

Analytical solutions of equations are of fundamental importance in understanding and describing physical phenomena, since they are able to take into account all the parameters of a problem, and investigate their influence and it easy to obtain the asymptotic behavior of the solution, which is usually difficult to generate through numerical calculations. Moreover, when using models, while they are rather sophisticated instruments that ultimately reflect the current state of knowledge on turbulent transport in the atmosphere, the results they provide are subject to a considerable margin of error. This is due to various factors, including in particular the uncertainty of the intrinsic variability of the atmosphere. Models, in fact, provide values expressed as an average, i.e. a mean value obtained by the repeated performance of many experiments, while the measured concentrations are a single value of the sample to which the ensemble average provided by models refer. This is a general characteristic of the theory of atmospheric turbulence and is a consequence of the statistical approach used in attempting to parameterize the chaotic character of the measured data. An analytical solution can be useful in evaluating the performances of numerical model (that solve numerically the advection diffusion equation) that could compare their results, not only against experimental data but, in an easier way, with the solution itself in order to check numerical errors without the uncertainties presented above.

Many operative models (using an analytical formula for the air pollution concentration) adopt empirical algorithms for describing dry deposition. The Gaussian plume equation was modified to include source depletion models (Chamberlain 1953; Overcamp 1976) and surface depletion models algorithms (Horst 1977, 1984). The solution proposed by Smith (1962), Ermak (1977), Rao (1981) also retained the framework of invariant wind speed and eddies with height (as the Gaussian approach). More recently, analytical solutions of advection-diffusion equation with dry deposition at the ground have utilized height-dependent wind speed and eddy diffusivities (Horst and Slinn 1984; Koch 1989; Chrysikopoulos et al. 1992; Lin and Hildemann 1997). However, these solutions are restricted to the specific case

where the source is located at the ground level and/or with restrictions to the wind speed and eddy diffusivities vertical profiles.

In this work we step forward solving analytically the two-dimensional, unsteady advection-diffusion-deposition equation using the GILTT (Generalized Integral Laplace Transform Technique) method. For more details about the methodology see the works of Wortmann et al. (2005), Moreira et al. (2005) and Moreira et al. (2006).

The dry deposition is described with a boundary condition of non-zero flux to the ground and without any restriction to the above profiles and the source position. Indeed, for this type of problem, the eigenvalues and eigenfunctions of the auxiliary Sturm-Liouville problem must be determined assuming boundary conditions of third type, which encompass the contaminant deposition speed. At this point it is worth noting that the mentioned works (Wortmann et al. 2005; Moreira et al. 2005; Moreira et al. 2006) assume boundary conditions only of second type.

To validate the results obtained, numerical comparison is undertaken with available results in the literature.

2. THE ANALYTICAL SOLUTION

For a Cartesian coordinate system in which the x direction coincides with that of the average wind, the unsteady two-dimension advection-diffusion equation with dry deposition to the ground is written as:

$$\frac{\partial c^y(x, z, t)}{\partial t} + u(z) \frac{\partial c^y(x, z, t)}{\partial x} = \frac{\partial}{\partial z} \left(K_z(z) \frac{\partial c^y(x, z, t)}{\partial z} \right) \quad (1)$$

subjected to the boundary conditions:

$$K_z(z) \frac{\partial c^y(x, z, t)}{\partial z} = V_g C(x, z, t) \quad \text{at } z = 0 \quad (1a)$$

$$K_z(z) \frac{\partial c^y(x, z, t)}{\partial z} = 0 \quad \text{at } z = h \quad (1b)$$

a continuous source condition:

$$u(z) c^y(0, z, t) = Q \delta(z - H_s) \quad \text{at } x = 0 \quad (1c)$$

and the initial condition:

$$c^y(x, z, 0) = 0 \quad \text{at } t = 0 \quad (1d)$$

Here, c^y denotes the pollutant concentration, K_z is the turbulent eddy diffusivity coefficient assumed to be a function of the variable z , u is the mean wind oriented in the x direction and function of the variable z , V_g the deposition velocity, h is the height of PBL, Q the emission rate, H_s the height of the source and δ is the Dirac-Delta function.

Using the Laplace Transform technique, transforming t into r and c^y into C , the equation (1) becomes:

$$u(z) \frac{\partial C(x, z, r)}{\partial x} = \frac{\partial}{\partial z} \left(K_z(z) \frac{\partial C(x, z, r)}{\partial z} \right) - rC(x, z, r) \quad (2)$$

To solve the problem by the GILTT method, Eq. (2) is rewritten as:

$$u(z) \frac{\partial C(x, z, r)}{\partial x} = K_z(z) \frac{\partial^2 C(x, z, r)}{\partial z^2} + K'_z(z) \frac{\partial C(x, z, r)}{\partial z} - rC(x, z, r) \quad (3)$$

where it should be noted that the first term on the right hand side satisfies the following Sturm-Liouville problem:

$$\zeta''_i(z) + \lambda_i^2 \zeta_i(z) = 0 \quad \text{at } 0 < z < h \quad (4)$$

$$-K_z \zeta'_i(z) + V_g \zeta_i(z) = 0 \quad \text{at } z = 0 \quad (4a)$$

$$\zeta'_i(z) = 0 \quad \text{at } z = h \quad (4b)$$

The solution of problem (3) constitutes a well known set of orthogonal eigenfunctions $\zeta_i(z) = \cos(\lambda_i(h-z))$ whose eigenvalues fulfill the ensuing transcendental equation:

$$\lambda_i(z) \tan(\lambda_i(z)h) = H_1 \quad (4c)$$

where $H_1 = \frac{V_g}{K_z}$. The eigenvalues are calculated solving the transcendental equation by the Newton-Raphson method.

It is now possible to apply the GILTT approach. For this purpose, the pollutant concentration is expanded in the serie:

$$C(x, z, r) = \sum_{i=0}^{\infty} \bar{c}_i(x, r) \zeta_i(z) \quad (5)$$

Replacing the above equation in Eq. (3) and taking moments, the following is obtained:

$$\begin{aligned} \sum_{i=0}^{\infty} \bar{c}_i(x, r) \int_0^h K'_z(z) \Psi'_i(z) \Psi_j(z) dz - \sum_{i=0}^{\infty} \lambda_i^2 \bar{c}_i(x, r) \int_0^h K_z(z) \Psi_i(z) \Psi_j(z) dz - \\ - \sum_{i=0}^{\infty} \bar{c}'_i(x, r) \int_0^h u(z) \Psi_i(z) \Psi_j(z) dz - \sum_{i=0}^{\infty} r \bar{c}_i(x, r) \int_0^h \Psi_i(z) \Psi_j(z) dz = 0 \end{aligned} \quad (6)$$

The above equation can be written in matrix fashion as:

$$Y'(x, r) + F.Y(x, r) = 0 \quad (7)$$

where $Y(x, r)$ is the column vector whose components are $\bar{c}_i(x, r)$, the matrix F is defined as $F = B^{-1}E$ and the matrices B and E are given by:

$$b_{i,j} = - \int_0^h u(z) \zeta_i(z) \zeta_j(z) dz \quad (8a)$$

and

$$e_{i,j} = \int_0^h K'_z(z) \Psi'_i(z) \Psi_j(z) dz - \lambda_i^2 \int_0^h K_z(z) \Psi_i(z) \Psi_j(z) dz - r \int_0^h \Psi_i(z) \Psi_j(z) dz \quad (8b)$$

Following the procedure of Wortmann et al. (2005), Moreira et al. (2005) and Moreira et al. (2006), one obtains the following solution for problem (7):

$$Y(x, r) = X.G(x, r).\xi \quad (9)$$

where X is the eigenfunction matrix of F , G is the diagonal matrix whose entries have the form $e^{-d_i x}$, d_i are the eigenvalues of F and ξ the vector given by $\xi = X^{-1}Y(0)$. Knowing the coefficients of the concentration series expansion, the solution for pollutant concentration given by Eq. (5) is well determined:

$$C(x, z, r) = \sum_{i=0}^{\infty} \bar{c}_i(x, r) \zeta_i(z) \quad (10)$$

where $\bar{c}_i(x, r)$ is the solution of the transformed problem given by Eq. (9), and $\zeta_i(z)$ comes from the solution of the Sturm-Liouville problem given in problem (4), where $\zeta_i(z) = \cos(\lambda_i(h-z))$.

Finally, the time-dependent concentration is obtained inverting numerically the transformed concentration $C(x, z, r)$ by the FT algorithm (Valkó and Abate, 2004; Abate and Valkó, 2004):

$$c(x, z, t) = \sum_{i=0}^{\infty} \Psi_i(z) \left(\frac{r}{M} \left\{ \frac{1}{2} \bar{c}_i(x, r) e^{rt} + \sum_{k=1}^{M-1} \text{Re} \left[e^{rS(\theta_k)} \bar{c}_i(x, S(\theta_k)) (1 + i\sigma(\theta_k)) \right] \right\} \right) \quad (11)$$

where $S(\theta_k) = r\theta(\cot\theta + i)$, $-\pi < \theta < +\pi$, $\sigma(\theta_k) = \theta_k + (\theta_k \cot\theta_k - 1)\cot\theta_k$, $\theta_k = \frac{k\pi}{M}$ and r is a parameter based

on numerical experiments. To control the round-off error in the computation of (11), we specify the precision requirement: number of precision decimal digits = M . No approximations are made in the derivation of this solution and so, it is analytical except for the round-off error and numerical inversion of time. The infinite series given in Eq. (11) can be truncated when the convergence is under a prefixed value. In the present case, 60 terms were utilized with an error of 0.5%.

3. EXPERIMENTAL DATA AND PBL PARAMETERIZATION

In order to show an example of the application of the obtained solution (Eq. (10)), the dataset of the Hanford diffusion experiment was used. This experiment was conducted in May-June, 1983 on a semi-arid region of south eastern Washington on generally flat terrain. The detailed description of the experiment was provided by Doran and Horst (1985). Data were obtained from six dual-tracer releases located at 100, 200, 800, 1600 and 3200m from the source during moderately stable to near-neutral conditions. However, the deposition velocity was evaluated only for the last 3 distances. The release time was 30 min except in run five, when it was 22 min. The terrain roughness was 3cm.

Two tracers, one depositing and one non-depositing, were released simultaneously from a height of 2 m. Zinc sulfide (ZnS) was chosen for the depositing tracer, while sulfur hexafluoride (SF₆) was the non-depositing tracer. The lateral separation between the SF₆ and ZnS release points was less than 1 m. The near-surface release height and the atmospheric stability conditions were chosen to produce differences between the depositing and non-depositing tracer concentrations that could be easily measured. The data collected during the field tests were tabulated (as crosswind-integrated tracer concentration data) and presented in Doran et al. (1984). The meteorological data and crosswind-integrated tracer concentration data, normalized by the release rate Q , are listed in Tab. 1. Note that in Tab. 1, C_d and C_{nd} are, respectively, the crosswind-integrated concentrations of ZnS and SF₆ normalized by the emission rate Q . For more details about the way that the effective deposition velocities and wind speed are calculated, also about the way that the measurements were taken, see the work of Doran and Horst (1985).

In order to use the above solution (Eq. (10)), it was necessary to select wind and eddy coefficient vertical profiles. The reliability of each model strongly depends on the way that turbulent parameters are calculated and related to the current understanding of the PBL (Seinfeld and Pandis, 1997).

The vertical eddy diffusivity used in this work is given in Degrazia et al. (2000):

$$K_z = \frac{0.3(1-z/h)u_*z}{1+3.7z/\Lambda} \quad (12)$$

where z is the height, u_* is the vertical convective velocity scale, $\Lambda = L(1-z/h)^{5/4}$ and L is the Monin-Obukhov length.

The wind velocity profile was described by a power law expressed as follows (Panofsky and Dutton 1988):

$$\frac{u_z}{u_1} = \left(\frac{z}{z_1} \right)^n \quad (13)$$

where u_z and u_1 are the mean wind velocity at the heights z and z_1 , while n is an exponent that is related to the intensity of turbulence for rural terrain (Irwin 1979).

Table 1. Tracer and meteorological data for six dual-tracer releases. L (m), u_* (cm.s^{-1}) and h (m) are the Monin-Obukhov length scale, the friction velocity and the PBL height, respectively, u is the wind velocity and V_g the deposition velocity. Subscript d refers to depositing material and subscript nd refers to non-depositing material.

Exp.	Arc (m)	ZnS/ Q (s.m^{-2})	SF ₆ / Q (s.m^{-2})	u (m.s^{-1})	V_g (cm.s^{-1})	C_d/C_{nd}
$u_* = 40$	800	0.00224	0.00373	7.61	4.21	0.601
$L = 166$	1600	0.000982	0.00214	8.53	4.05	0.459
$h = 325$	3200	0.000586	0.00130	9.43	3.65	0.451
$u_* = 26$	800	0.00747	0.0129	3.23	1.93	0.579
$L = 44$	1600	0.00325	0.00908	3.59	1.80	0.358
$h = 135$	3200	0.00231	0.00722	3.83	1.74	0.320
$u_* = 27$	800	0.00306	0.00591	4.74	3.14	0.518
$L = 77$	1600	0.00132	0.00331	5.40	3.02	0.399
$h = 182$	3200	0.000662	0.00179	6.32	2.84	0.370
$u_* = 20$	800	0.00804	0.0201	3.00	1.75	0.400
$L = 34$	1600	0.00426	0.0131	3.39	1.62	0.325
$h = 104$	3200	0.00314	0.00915	3.75	1.31	0.343
$u_* = 26$	800	0.00525	0.0105	3.07	1.56	0.500
$L = 59$	1600	0.00338	0.00861	3.24	1.47	0.393
$h = 157$	3200	0.00292	0.00664	3.46	1.14	0.440
$u_* = 30$	800	0.00723	0.0134	3.17	1.17	0.540
$L = 71$	1600	0.00252	0.00615	3.80	1.15	0.410
$h = 185$	3200	0.00125	0.00311	4.37	1.10	0.402

4. NUMERICAL RESULTS

The model was evaluated with the ratio C_d/C_{nd} , where C_d and C_{nd} are the crosswind-integrated concentrations of ZnS and SF₆ measured at 1.5 m above the ground and normalized respectively by the emission rate Q . A comparison of predicted and observed values C_d/C_{nd} are shown in Fig. 1 for approach (11), with vertical eddy diffusivity given by Degrazia et al. (2000) and power profile of wind (Panofsky and Dutton 1988). Data between dot lines correspond to a factor of two. In this respect, it is possible to note that the model reproduces fairly well the observed concentration.

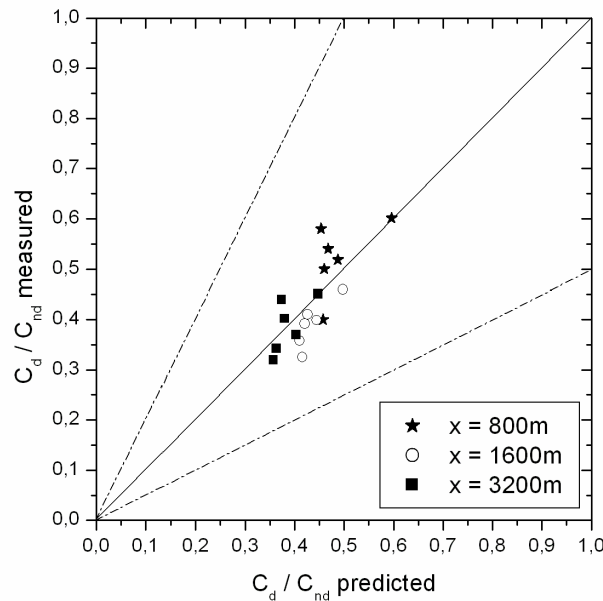


Figure 1. Scatter diagram of observed and predicted data. Data between dot lines correspond to a factor of two.

Table 2 presents some performance measurements, obtained using the well known statistical evaluation procedure described by Hanna (1989):

$$\text{Normalized mean square error (NMSE)} = \overline{(C_o - C_p)^2} / \overline{C_o C_p},$$

$$\text{Factor of due (FA2)} = \text{fraction of data (\%)} \text{ for } 0.5 \leq (C_p / C_o) \leq 2$$

$$\text{Correlation coefficient (COR)} = \overline{(C_o - \overline{C_o})(C_p - \overline{C_p})} / \sigma_o \sigma_p,$$

$$\text{Fractional bias (FB)} = \overline{C_o} - \overline{C_p} / 0.5(\overline{C_o} + \overline{C_p}),$$

$$\text{Fractional standard deviations (FS)} = (\sigma_o - \sigma_p) / 0.5(\sigma_o + \sigma_p)$$

where subscripts o and p refer to observed and predicted quantities, respectively, σ is the standard deviation and an overbar indicates an average.

Table 2. Statistical evaluation of model performance.

	NMSE	COR	FA2	FB	FS
GILTT	0.01	0.77	1.00	-0.01	0.38

The statistical index FB indicates whether the predicted quantities underestimate or overestimate the observed ones. The statistical index $NMSE$ represents the quadratic error of the predicted quantities in relation to the observed ones. Best results are indicated by values nearest zero in $NMSE$, FB and FS , and nearest 1 in R and $FA2$. The statistical indices point out that a good agreement is obtained between experimental data and the GILTT model. The computational time to obtain the numerical results was 72 seconds in an Intel Celeron, 1.60GHz and 1024Mb of RAM.

Doran and Horst (1985) presented four different models that evaluate the dry deposition at the ground with four different approaches: the source depletion approach of Chamberlain (1953), the corrected source depletion model of Horst (1980, 1983), the K model proposed by Ermak (1977) and Rao (1981), and the K corrected model of Rao (1981). Finally, to compare the results with the four models above, different statistical parameters were calculated (used in the paper by Doran and Horst, 1985) described by Fox (1981) and Willmott (1982):

$$\text{Mean bias } (\overline{d}) = \sum_{i=1}^N d_i / N$$

$$\text{Variance } (S^2) = \sum_{i=1}^N (d_i - \overline{d})^2 / (N - 1)$$

$$\text{Mean absolute error (MAE)} = \sum_{i=1}^N |Cp_i - Co_i| / N$$

$$\text{Index of agreement (I)} = 1 - \left[\frac{\sum_{i=1}^N (P_i' - O_i')^2}{\sum_{i=1}^N (|P_i'| + |O_i'|)^2} \right]$$

where d_i is the difference between observed (Co_i) and predicted (Cp_i) values, $P_i' = Cp_i - \overline{Co_i}$, $O_i' = Co_i - \overline{Co_i}$, the overbar indicates an average and $0 < I < 1$ and N is the data number.

In Tab. 3 comparisons between the GILTT approach and the above models (Chamberlain, 1953; Horst, 1980; Horst, 1983; Ermak, 1977; Rao, 1981) are reported, and it is possible to see the good performance of the solution.

Table 3. Statistical evaluation of model performance compared with other models.

Parameter	GILTT	Source depletion	Corrected source depletion	K model	Corrected K model
Mean Bias	0.01	0.11	0.01	0.21	0.07
Mean absolute error	0.04	0.11	0.05	0.21	0.07
S = (variance) ^{1/2}	0.05	0.05	0.06	0.08	0.05
Correlation coefficient	0.77	0.82	0.70	0.63	0.78
Index of agreement	0.84	0.64	0.83	0.42	0.76

5. FINAL REMARKS

A general solution of the two-dimension time-dependent advection-diffusion equation considering dry deposition to the ground has been presented. In order to show the performances of the solution in actual scenarios, a parameterization of the PBL has been introduced, and the values predicted by the solutions have been compared with the Hanford diffusion experiment dataset. The analysis of the results shows a reasonably good agreement between the computed values against the experimental ones. Finally, the solution results were compared with those of 4 different models.

Therefore the methodology discussed is promising to simulate pollutant dispersion in atmosphere. Furthermore, the use of the FT algorithm allows us to obtain results with a prescribed accuracy.

We focus our future attention in the task of improving this methodology in order to make it more operational for air quality modeling.

6. ACKNOWLEDGEMENTS

The authors thank to CNPq (Conselho Nacional de Desenvolvimento Científico e Tecnológico), CAPES (Coordenação de Aperfeiçoamento de Pessoal de Nível Superior) and the project “Laboratorio LaRIA” for the partial financial support of this work.

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